

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

Attorney's Docket Number  
05725.0555

U.S. Application No.

09/486558

|                               |                           |                       |
|-------------------------------|---------------------------|-----------------------|
| International Application No. | International Filing Date | Priority Date Claimed |
| PCT/FR98/01591                | July 20, 1998             | September 1, 1997     |

Title of Invention:

DYEING COMPOSITION FOR KERATIN FIBRES

430 Rec'd PCT/PTO 29 FEB 2000

Applicant(s) For DO/EO/US:

Mireille MAUBRU and Marie-Pascale AUDOUSSET

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [X] This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. [ ] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. [ ] This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. [X] A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. [X] A copy of the International Application as filed (35 U.S.C. 371(c)(2))
- a. [ ] is transmitted herewith (required only if not transmitted by the International Bureau).
- b. [X] has been transmitted by the International Bureau.
- c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).
6. [X] A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. [X] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
- a. [ ] are transmitted herewith (required only if not transmitted by the International Bureau).
- b. [ ] have been transmitted by the International Bureau.
- c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
- d. [X] have not been made and will not be made.
8. [ ] A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. [ ] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. [ ] A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. [X] An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. [ ] An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. [ ] A FIRST preliminary amendment.
- [ ] A SECOND or SUBSEQUENT preliminary amendment.
14. [ ] A substitute specification.
15. [ ] A change of power of attorney and/or address letter.
16. [ ] Other items or information:
- a. [ ] Verified Small Entity Statement.
- b. [ ] Copy of Notification of Missing Requirements.

09/486558

PCT/FR98/01591

430 Rec'd PCT/TO 05/25/0555

29 FEB 2000

17. [X] The following fees are submitted:

CALCULATIONS

**Basic National Fee (37 CFR 1.492(a)(1)-(5)):**

Search Report has been prepared by the EPO or JPO.....\$840.00  
 International preliminary examination fee paid to  
 USPTO (37 CFR 1.482).....\$670.00  
 No international preliminary examination fee paid to  
 USPTO (37 CFR 1.482) but international search fee  
 paid to USPTO (37 CFR 1.445(a)(2)).....\$760.00  
 Neither international preliminary examination fee  
 (37 CFR 1.482) nor international search fee  
 (37 CFR 1.445(a)(2)) paid to USPTO.....\$970.00  
 International preliminary examination fee paid to USPTO  
 (37 CFR 1.482) and all claims satisfied provisions  
 of PCT Article 33(1)-(4).....\$ 96.00

|   |          |                  |
|---|----------|------------------|
| <b>ENTER APPROPRIATE BASIC FEE AMOUNT</b> | <b>=</b> | <b>\$ 840.00</b> |
|---|----------|------------------|

Surcharge of \$130.00 for furnishing the oath or declaration later than  
 [ ] 20 [ ] 30 months from the earliest claimed priority date  
 (37 CFR 1.492(e)).

| Claims                                      | Number Filed | Number Extra | Rate      |                     |
|---|--------------|--------------|-----------|---------------------|
| Total Claims                                | 25 -20=      | 5            | X \$18.00 | \$ 90.00            |
| Independent Claims                          | 1 - 3=       |              | X \$78.00 | \$                  |
| Multiple dependent claim(s) (if applicable) |              |              | +\$260.00 | \$ 260.00           |
| <b>TOTAL OF ABOVE CALCULATIONS</b>          |              |              |           | <b>= \$1,190.00</b> |

Reduction by 1/2 for filing by small entity, if applicable. Verified  
 Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28)

|                 |          |                   |
|-----------------|----------|-------------------|
| <b>SUBTOTAL</b> | <b>=</b> | <b>\$1,190.00</b> |
|-----------------|----------|-------------------|

Processing fee of \$130.00 for furnishing the English translation later  
 than [ ] 20 [ ] 30 months from the earliest claimed priority date  
 (37 CFR 1.492(f)).

|                           |          |                   |
|---------------------------|----------|-------------------|
| <b>TOTAL NATIONAL FEE</b> | <b>=</b> | <b>\$1,190.00</b> |
|---------------------------|----------|-------------------|

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The  
 assignment must be accompanied by an appropriate cover sheet  
 (37 CFR 3.28, 3.31).


|                            |          |                   |
|----------------------------|----------|-------------------|
| \$40.00 per property       | +        | \$                |
| <b>TOTAL FEES ENCLOSED</b> | <b>=</b> | <b>\$1,190.00</b> |

|              |    |
|--------------|----|
| Amount to be |    |
| refunded     | \$ |
| charged      | \$ |

- a. [X] A check in the amount of \$1,190.00 to cover the above fees is enclosed.  
 b. [ ] Please charge my Deposit Account No. \_\_\_\_\_ in the amount of  
 \$\_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is  
 enclosed.  
 c. [X] The Commissioner is hereby authorized to charge any additional fees  
 which may be required, or credit any overpayment to Deposit Account  
 No. 06-0916. A duplicate copy of this sheet is enclosed.

The Commissioner is hereby authorized to charge any other fees due under 37 C.F.R. §1.16  
 or §1.17 during the pendency of this application to our Deposit Account No. 06-0916.

SEND ALL CORRESPONDENCE TO:  
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 Ernest F. Chapman  
 Reg. No. 25,961

Submitted: February 29, 2000

09/486558



13 APR 2000

PATENT

Attorney Docket No.: 5725.0555-00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re National Stage of International )  
Application No. PCT/FR98/01591 of: )  
Mireille MAUBRU et al. )  
Serial No.: 09/486,558 ) Group Art Unit: Unassigned  
PCT Filed: July 20, 1998 ) Examiner: Unassigned  
National Stage Entry: February 29, 2000 )  
FOR: DYEING COMPOSITION FOR )  
KERATIN FIBRES )

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231

Attention: BOX PCT

Sir:

Prior to the examination of the above application, please amend the abstract,  
cancel claims 1 to 19 without prejudice or disclaimer and add new claims 20 to 47  
as follows:

IN THE ABSTRACT:

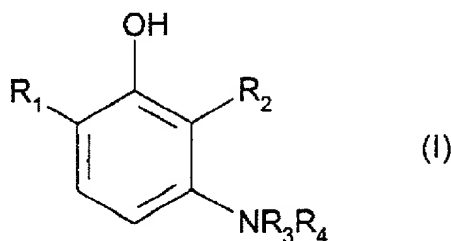
Please delete the abstract and insert the new abstract, which is attached on  
a separate sheet.

09/486558-04300

**IN THE CLAIMS:**

--20. A composition for the oxidation dyeing of keratin fibers comprising:

- at least one oxidation base chosen from diaminopyrazoles, triaminopyrazoles, and acid-addition salts thereof;
- and at least one coupler chosen from halogenated meta-aminophenols of formula (I), and acid addition salts thereof:



in which:

- R<sub>1</sub> and R<sub>2</sub>, which are identical or different, are chosen from a hydrogen atom, a halogen atom, a C<sub>1</sub>-C<sub>4</sub> alkyl radical, a C<sub>1</sub>-C<sub>4</sub> monohydroxyalkyl radical, a C<sub>2</sub>-C<sub>4</sub> polyhydroxyalkyl radical, a C<sub>1</sub>-C<sub>4</sub> alkoxy radical, a C<sub>1</sub>-C<sub>4</sub> monohydroxyalkoxy radical and a C<sub>2</sub>-C<sub>4</sub> polyhydroxyalkoxy radical;

- R<sub>3</sub> and R<sub>4</sub>, which are identical or different, are chosen from a hydrogen atom, a C<sub>1</sub>-C<sub>4</sub> alkyl radical, a C<sub>1</sub>-C<sub>4</sub> monohydroxyalkyl radical, a C<sub>2</sub>-C<sub>4</sub> polyhydroxyalkyl radical and a C<sub>1</sub>-C<sub>4</sub> monoaminoalkyl radical;

with the proviso that at least one of said radicals R<sub>1</sub> and R<sub>2</sub> is a halogen atom.

21. A composition according to Claim 20, wherein said keratin fibers are human keratin fibers.

22. A composition according to Claim 21, wherein said human keratin fibers are human hair.

23. A composition according to Claim 20, wherein said composition is in a medium suitable for dyeing.

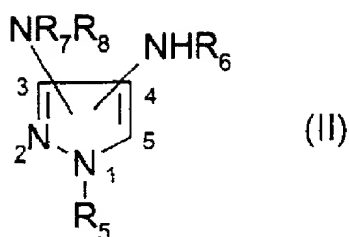
24. A composition according to Claim 20, wherein said halogen atoms are chosen from chlorine, bromine, iodine and fluorine.

25. A composition according to Claim 20, wherein said halogenated meta-aminophenols of formula (I) are chosen from 3-amino-6-chlorophenol, 3-amino-6-bromophenol, 3-(β-aminoethyl)amino-6-chlorophenol, 3-(β-hydroxyethyl)amino-6-chlorophenol and 3-amino-2-chloro-6-methylphenol, and acid addition salts thereof.

26. A composition according to Claim 20, wherein said diaminopyrazoles are chosen from:

a) diaminopyrazoles of formula (II), and acid addition salts thereof:

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in which:

-  $R_5$  is chosen from a hydrogen atom, a  $C_1$ - $C_6$  alkyl radical, a  $C_2$ - $C_4$  hydroxyalkyl radical, a benzyl radical, a phenyl radical, a benzyl radical substituted with a halogen atom, a  $C_1$ - $C_4$  alkyl radical or  $C_1$ - $C_4$  alkoxy radical, or

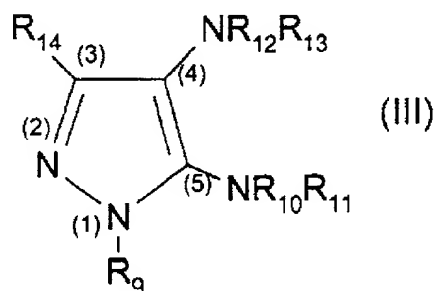
$R_5$  forms, with the nitrogen atom of the group  $NR_7R_8$  in position 5, a hexahydropyridazine or tetrahydropyrazole heterocycle which is optionally monosubstituted with a  $C_1$ - $C_4$  alkyl group;

-  $R_6$  and  $R_7$  which are identical or different, are chosen from a hydrogen atom, a  $C_1$ - $C_4$  alkyl radical, a  $C_2$ - $C_4$  hydroxyalkyl radical, a benzyl radical and a phenyl radical;

-  $R_8$  is chosen from a hydrogen atom, a  $C_1$ - $C_6$  alkyl radical and a  $C_2$ - $C_4$  hydroxyalkyl radical;

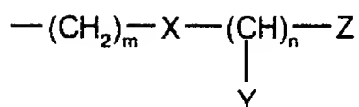
with the proviso that  $R_6$  is a hydrogen atom when  $R_5$  either is a substituted benzyl radical or forms a heterocycle with the nitrogen atom of the group  $NR_7R_8$  in position 5; and

b) diaminopyrazoles of formula (III), and acid addition salts thereof:



in which:

-  $R_9$ ,  $R_{10}$ ,  $R_{11}$ ,  $R_{12}$  and  $R_{13}$ , which are identical or different, are chosen from a hydrogen atom; a linear or branched  $C_1$ - $C_6$  alkyl radical; a  $C_2$ - $C_4$  hydroxyalkyl radical; a  $C_2$ - $C_4$  aminoalkyl radical; a phenyl radical; a phenyl radical substituted with a halogen atom or a  $C_1$ - $C_4$  alkyl,  $C_1$ - $C_4$  alkoxy, nitro, trifluoromethyl, amino or  $C_1$ - $C_4$  alkylamino radical; a benzyl radical; a benzyl radical substituted with a halogen atom or with a  $C_1$ - $C_4$  alkyl,  $C_1$ - $C_4$  alkoxy, methylenedioxy or amino radical; and a radical



in which m and n are integers, which are identical or different, ranging from 1 to 3 inclusive, X is chosen from an oxygen atom and an NH group, Y is chosen from a hydrogen atom and a methyl radical, and Z is chosen from a methyl radical and a group OR or NRR' in which R and R', which are identical or different, are chosen from a hydrogen atom, a methyl radical and an ethyl radical, with the proviso that when R<sub>10</sub> is a hydrogen atom, then R<sub>11</sub> can also be an amino or C<sub>1</sub>-C<sub>4</sub> alkylamino radical,

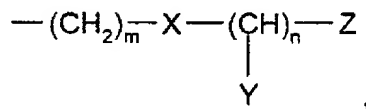
- R<sub>14</sub> is chosen from a linear or branched C<sub>1</sub>-C<sub>6</sub> alkyl radical; a C<sub>1</sub>-C<sub>4</sub> hydroxyalkyl radical; a C<sub>1</sub>-C<sub>4</sub> aminoalkyl radical; a (C<sub>1</sub>-C<sub>4</sub>)alkylamino(C<sub>1</sub>-C<sub>4</sub>)alkyl radical; a di(C<sub>1</sub>-C<sub>4</sub>)alkylamino(C<sub>1</sub>-C<sub>4</sub>)alkyl radical; a hydroxy(C<sub>1</sub>-C<sub>4</sub>)alkylamino(C<sub>1</sub>-C<sub>4</sub>)alkyl radical; a (C<sub>1</sub>-C<sub>4</sub>)alkoxymethyl radical; a phenyl radical; a phenyl radical substituted with a halogen atom or with a C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy, nitro, trifluoromethyl, amino or C<sub>1</sub>-C<sub>4</sub> alkylamino radical; a benzyl radical; a benzyl radical substituted with a halogen atom or with a C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy, nitro, trifluoromethyl, amino or C<sub>1</sub>-C<sub>4</sub> alkylamino radical; a heterocycle chosen from thiophene, furan and pyridine; and a radical -(CH<sub>2</sub>)<sub>p</sub>-O-(CH<sub>2</sub>)<sub>q</sub>-OR", in which p and q are integers, which are identical or



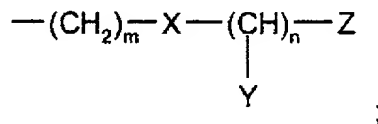
different, ranging from 1 to 3 inclusive, and R" is chosen from a hydrogen atom and a methyl radical;

with the provisos that, in formula (III),

- at least one of the radicals R<sub>10</sub>, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub> is a hydrogen atom;
- when R<sub>10</sub>, or R<sub>12</sub>, respectively, is a substituted or unsubstituted phenyl radical, or a benzyl radical or a radical



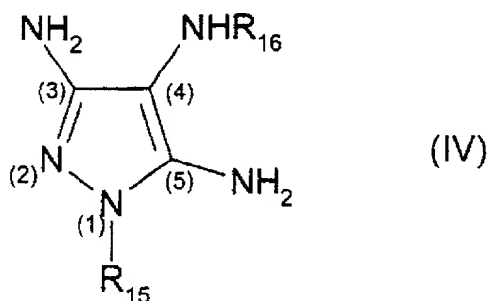
then R<sub>11</sub>, or R<sub>13</sub>, respectively, is not a substituted or unsubstituted phenyl radical, or a benzyl radical or a radical



- when R<sub>12</sub> and R<sub>13</sub> simultaneously represent a hydrogen atom, then R<sub>9</sub> can form, with R<sub>10</sub> and R<sub>11</sub>, a hexahydropyrimidine or tetrahydroimidazole heterocycle which is optionally substituted with a C<sub>1</sub>-C<sub>4</sub> alkyl or 1,2,4-tetrazole radical;
- when R<sub>10</sub>, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub> represent a hydrogen atom or a C<sub>1</sub>-C<sub>6</sub> alkyl radical, then R<sub>9</sub> or R<sub>14</sub> can also represent a 2-, 3- or 4-pyridyl, 2- or 3-thienyl or 2- or 3-furyl

heterocyclic residue which is optionally substituted with a methyl radical or a cyclohexyl radical.

27. A composition according to Claim 20, wherein said triaminopyrazoles are chosen from compounds of formula (IV), and acid addition salts thereof:



in which:

-  $R_{15}$  and  $R_{16}$ , which are identical or different, are chosen from a hydrogen atom, a  $C_1$ - $C_4$  alkyl and a  $C_2$ - $C_4$  hydroxyalkyl radical.

28. A composition according to Claim 26, wherein said diaminopyrazoles of formula (II) are chosen from:

- 4,5-diamino-1-(4'-methoxybenzyl)pyrazole,
- 4,5-diamino-1-(4'-methylbenzyl)pyrazole,

- 4,5-diamino-1-(4'-chlorobenzyl)pyrazole,
  - 4,5-diamino-1-(3'-methoxybenzyl)pyrazole,
  - 4-amino-1-(4'-methoxybenzyl)-5-methylaminopyrazole,
  - 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(4'-methoxybenzyl)pyrazole,
  - 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-methylpyrazole,
  - 4-amino-(3)5-methylaminopyrazole,
  - 3-(5)4-diaminopyrazole,
  - 4,5-diamino-1-methylpyrazole,
  - 4,5-diamino-1-benzylpyrazole,
  - 3-amino-4,5,7,8-tetrahydropyrazolo{1,5-a}pyrimidine,
  - 7-amino-2,3-dihydro-1H-imidazolo{1,2-b}pyrazole,
  - 3-amino-8-methyl-4,5,7,8-tetrahydropyrazolo{1,5-a}pyrimidine,
- and acid addition salts thereof.

29. A composition according to Claim 26, wherein said diaminopyrazoles of formula (III) are chosen from:

- 1-benzyl-4,5-diamino-3-methylpyrazole,
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(4'-methoxyphenyl)pyrazole,
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(4'-methylphenyl)pyrazole,
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(3'-methylphenyl)pyrazole,
- 4,5-diamino-3-methyl-1-isopropylpyrazole,

- 10 -

- 4,5-diamino-3-dimethylaminomethyl-1-tert-butylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-ethylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-isopropylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-tert-butylpyrazole,
- 4,5-diamino-3-methylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-methylaminomethyl-1-isopropylpyrazole,
- 4,5-diamino-1-ethyl-3-methylaminomethylpyrazole,
- 1-tert-butyl-4,5-diamino-3-methylaminomethylpyrazole,
- 4,5-diamino-3- $\{(\beta\text{-hydroxyethyl})\text{aminomethyl}\}$ -1-methylpyrazole,
- 4,5-diamino-3- $\{(\beta\text{-hydroxyethyl})\text{aminomethyl}\}$ -1-isopropylpyrazole,
- 4,5-diamino-1-ethyl-3- $\{(\beta\text{-hydroxyethyl})\text{aminomethyl}\}$ pyrazole,
- 1-tert-butyl-4,5-diamino-3- $\{(\beta\text{-hydroxyethyl})\text{aminomethyl}\}$ pyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1,3-dimethylpyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1-isopropyl-3-methylpyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1-ethyl-3-methylpyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1-tert-butyl-3-methylpyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1-phenyl-3-methylpyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1-(2-methoxyphenyl)-3-methylpyrazole,
- 4-amino-5-( $\beta\text{-hydroxyethyl}$ )amino-1-(3-methoxyphenyl)-3-methylpyrazole,

- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(4-methoxyphenyl)-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-benzyl-3-methylpyrazole,
- 4-amino-1-ethyl-3-methyl-5-methylaminopyrazole,
- 4-amino-1-tert-butyl-3-methyl-5-methylaminopyrazole,
- 4,5-diamino-1,3-dimethylpyrazole,
- 4,5-diamino-3-tert-butyl-1-methylpyrazole,
- 4,5-diamino-1-tert-butyl-3-methylpyrazole,
- 4,5-diamino-1-methyl-3-phenylpyrazole,
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-methylpyrazole,
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-phenylpyrazole,
- 4,5-diamino-1-methyl-3-(2'-chlorophenyl)pyrazole,
- 4,5-diamino-1-methyl-3-(4'-chlorophenyl)pyrazole,
- 4,5-diamino-1-methyl-3-(3'-trifluoromethylphenyl)pyrazole,
- 4,5-diamino-1,3-diphenylpyrazole,
- 4,5-diamino-3-methyl-1-phenylpyrazole,
- 4-amino-1,3-dimethyl-5-phenylaminopyrazole,
- 4-amino-1-ethyl-3-methyl-5-phenylaminopyrazole,
- 4-amino-1,3-dimethyl-5-methylaminopyrazole,
- 4-amino-3-methyl-1-isopropyl-5-methylaminopyrazole,
- 4-amino-3-isobutoxymethyl-1-methyl-5-methylaminopyrazole,

- LAW OFFICES  
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202-408-4000

- 4-amino-5-(4'-chlorophenyl)amino-3-phenylpyrazole,
- 4-amino-3-(4'-chlorophenyl)-5-phenylaminopyrazole,
- 4-amino-3-(4'-methoxyphenyl)-5-phenylaminopyrazole,
- 1-(4'-chlorobenzyl)-4,5-diamino-3-methylpyrazole,
- 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole,
- 4-amino-1-ethyl-3-methyl-5-methylaminopyrazole,
- 4-amino-5-(2'-aminoethyl)amino-1,3-dimethylpyrazole,

and acid addition salts thereof.

30. A composition according to Claim 29, wherein said diaminopyrazoles of formula (III) are chosen from:

- 4,5-diamino-1,3-dimethylpyrazole,
- 4,5-diamino-3-methyl-1-phenylpyrazole,
- 4,5-diamino-1-methyl-3-phenylpyrazole,
- 4-amino-1,3-dimethyl-5-hydrazinopyrazole,
- 1-benzyl-4,5-diamino-3-methylpyrazole,
- 4,5-diamino-3-tert-butyl-1-methylpyrazole,
- 4,5-diamino-1-tert-butyl-3-methylpyrazole,
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-methylpyrazole,
- 4,5-diamino-1-ethyl-3-methylpyrazole,
- 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole,



- 4,5-diamino-1-ethyl-3-hydroxymethylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-methylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole,
  - 4,5-diamino-3-methyl-1-isopropylpyrazole,
  - 4-amino-5-(2'-aminoethyl)amino-1,3-dimethylpyrazole,
- and acid addition salts thereof.

31. A composition according to Claim 27 wherein said triaminopyrazoles of formula (IV) are chosen from 3,4,5-triaminopyrazole, 1-methyl-3,4,5-triaminopyrazole, 3,5-diamino-1-methyl-4-methylaminopyrazole and 3,5-diamino-4-( $\beta$ -hydroxyethyl)amino-1-methylpyrazole, and acid addition salts thereof.

32. A composition according to Claim 20, wherein said at least one oxidation base is present in an amount ranging from 0.0005 to 12% by weight relative to the total weight of the composition.

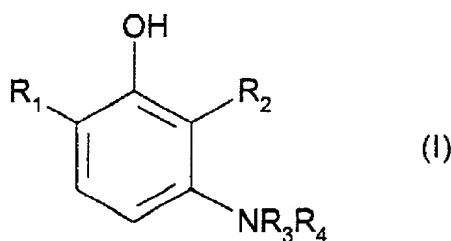
33. A composition according to Claim 32, wherein said at least one oxidation base is present in an amount ranging from 0.005 to 6% by weight relative to the total weight of the composition.

34. A composition according to Claim 20, wherein said at least one coupler is present in an amount ranging from 0.0001 to 5% by weight relative to the total weight of the composition.

42. A method for dyeing keratin fibers, comprising:

(a) applying to said keratin fibers at least one dye composition, which comprises

- at least one oxidation base chosen from diaminopyrazoles, triaminopyrazoles, and acid-addition salts thereof;
- and at least one coupler chosen from halogenated meta-aminophenols of formula (I), and acid addition salts thereof:



in which:

- $R_1$  and  $R_2$ , which are identical or different, are chosen from a hydrogen atom, a halogen atom, a  $C_1$ - $C_4$  alkyl radical, a  $C_1$ - $C_4$  monohydroxyalkyl radical, a  $C_2$ - $C_4$  polyhydroxyalkyl radical, a  $C_1$ - $C_4$  alkoxy radical, a  $C_1$ - $C_4$  monohydroxyalkoxy radical and a  $C_2$ - $C_4$  polyhydroxyalkoxy radical;

with the proviso that at least one of said radicals  $R_1$  and  $R_2$  is a halogen atom;  
and

43. A method according to Claim 42, wherein said keratin fibers are human keratin fibers.

44. A method according to Claim 43, wherein said human keratin fibers are human hair.

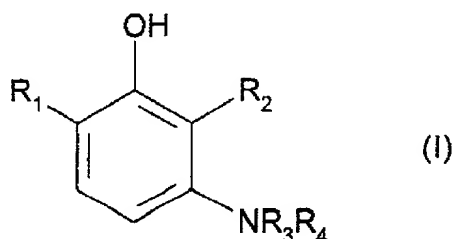
45. A method according to Claim 42, wherein said oxidizing agent is chosen from hydrogen peroxide, urea peroxide, alkali metal bromates, persalts, and peracids.

46. A method according to Claim 45, wherein said persalts are chosen from perborates, percarbonates and persulphates.

47. A multi-compartment kit for dyeing keratin fibers, comprising at least two compartments, wherein one compartment comprises an oxidizing composition,

and another compartment comprises a composition for the oxidation dyeing of keratin fibers, said composition for the oxidation dyeing of keratin fibers comprising:

- at least one oxidation base chosen from diaminopyrazoles, triaminopyrazoles, and acid-addition salts thereof;
- and at least one coupler chosen from halogenated meta-aminophenols of formula (I), and acid addition salts thereof:



in which:

-  $R_1$  and  $R_2$ , which are identical or different, are chosen from a hydrogen atom, a halogen atom, a  $C_1$ - $C_4$  alkyl radical, a  $C_1$ - $C_4$  monohydroxyalkyl radical, a  $C_2$ - $C_4$  polyhydroxyalkyl radical, a  $C_1$ - $C_4$  alkoxy radical, a  $C_1$ - $C_4$  monohydroxyalkoxy radical and a  $C_2$ - $C_4$  polyhydroxyalkoxy radical;

REMARKS

Please grant any extensions of time required to enter this Preliminary Amendment and charge any additional required fees to our deposit account Deposit Account No. 06-0916.

Respectfully submitted,

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Dated: April 13, 2000

## ABSTRACT

[illegible]

COMPOSITION FOR THE OXIDATION DYEING OF KERATIN FIBRES,  
COMPRISING A DIAMINOPYRAZOLE OR A TRIAMINOPYRAZOLE AND  
A HALOGENATED META-AMINOPHENOL, AND DYEING PROCESS

5           The present invention relates to a  
composition for the oxidation dyeing of keratin fibres,  
in particular human keratin fibres such as the hair,  
comprising at least one oxidation base chosen from  
diaminopyrazoles and triaminopyrazoles, in combination  
10 with at least one meta-aminophenol which is halogenated  
ortho to the phenol, as coupler, and to the dyeing  
process using this composition with an oxidizing agent.

It is known practice to dye keratin fibres,  
and in particular human hair, with dye compositions  
15 containing oxidation dye precursors, in particular  
ortho- or para-phenylenediamines, ortho- or para-  
aminophenols or heterocyclic compounds such as pyrazole  
derivatives, which are generally referred to as  
oxidation bases. Oxidation dye precursors, or oxidation  
20 bases, are colourless or weakly coloured compounds  
which, when combined with oxidizing products, can give  
rise to coloured compounds and dyes by means of a  
process of oxidative condensation.

It is also known that the shades obtained  
25 with oxidation bases can be varied by combining them  
with suitably selected couplers or coloration

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modifiers, the latter possibly being chosen in particular from aromatic meta-diamines, meta-aminophenols, meta-diphenols and certain heterocyclic compounds.

5           The variety of molecules used as oxidation bases and couplers allows a wide range of colours to be obtained.

          The so-called "permanent" coloration obtained by means of these oxidation dyes must moreover satisfy  
10 a certain number of requirements. Thus, it must have no toxicological drawbacks, it must allow shades to be obtained in the desired intensity and it must satisfactorily withstand external agents (light, bad weather, washing, permanent-waving, perspiration or  
15 rubbing).

          The dyes must also be able to cover white hair, and, lastly, they must be as unselective as possible, i.e. they must allow only the smallest possible differences in colour along the same keratin  
20 fibre, which may in fact be differently sensitized (i.e. damaged) between its tip and its root.

          . Compositions for the oxidation dyeing of keratin fibres, containing pyrazole derivatives such as 4,5-diaminopyrazoles, 3,4-diaminopyrazoles or 3,4,5-  
25 triaminopyrazoles as oxidation base, in combination with couplers conventionally used for oxidation dyeing,

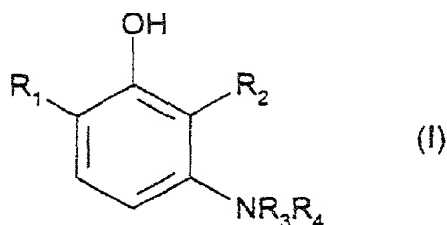
such as, for example, meta-phenylenediamines, meta-aminophenols, meta-diphenols and heterocyclic couplers such as, for example, indole derivatives, have already been proposed, in particular in German patent applications DE 3 843 892, DE 4 234 887, DE 4 234 886, DE 4 234 885 and DE 195 43 988. However, such compositions are not entirely satisfactory, in particular as regards the fastness of the colorations obtained with regard to the various attacking factors to which the hair may be subjected, and in particular with regard to perspiration.

However, the Applicant has now discovered that it is possible to obtain novel powerful dyes that are particularly resistant to the various attacking factors to which the hair may be subjected, by combining, as oxidation base, at least one diaminopyrazole and/or at least one triaminopyrazole and, as coupler, a meta-aminophenol halogenated in a position ortho to the phenol.

This discovery forms the basis of the present invention.

A first subject of the invention is thus a composition for the oxidation dyeing of keratin fibres and in particular human keratin fibres such as the hair, characterized in that it comprises, in a medium which is suitable for dyeing:

- at least one oxidation base chosen from diaminopyrazoles and triaminopyrazoles;
  - and at least one coupler chosen from the halogenated meta-aminophenols of formula (I) below, and the
- 5 addition salts thereof with an acid:



in which:

- $R_1$  and  $R_2$ , which may be identical or different, represent a hydrogen atom, a halogen atom such as
- 10 chlorine, bromine, iodine or fluorine, a  $C_1$ - $C_4$  alkyl radical, a  $C_1$ - $C_4$  monohydroxyalkyl radical, a  $C_2$ - $C_4$  polyhydroxyalkyl radical, a  $C_1$ - $C_4$  alkoxy radical, a  $C_1$ - $C_4$  monohydroxyalkoxy radical or a  $C_2$ - $C_4$  polyhydroxyalkoxy radical;
- 15 -  $R_3$  and  $R_4$ , which may be identical or different, represent a hydrogen atom, a  $C_1$ - $C_4$  alkyl radical, a  $C_1$ - $C_4$  monohydroxyalkyl radical, a  $C_2$ - $C_4$  polyhydroxyalkyl radical or a  $C_1$ - $C_4$  monoaminoalkyl radical;
- it being understood that at least one of the radicals  $R_1$
- 20 and  $R_2$  represents a halogen atom.

The oxidation dye composition in accordance with the invention makes it possible to obtain intense colorations in varied shades, which are relatively

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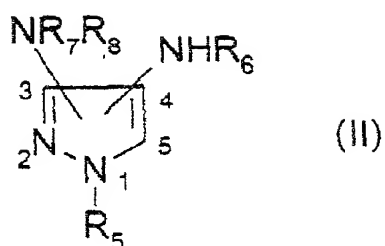
unselective and which have excellent properties of resistance both with respect to atmospheric agents such as light and bad weather, and with respect to perspiration and the various treatments to which the hair may be subjected (shampooing, permanent-waving). These properties are particularly noteworthy especially as regards the resistance of the colorations with respect to perspiration.

Among the  $C_1$ - $C_4$  alkyl and  $C_1$ - $C_4$  alkoxy radicals of the compounds of formula (I) above, mention may be made in particular of the methyl, ethyl, propyl, methoxy and ethoxy radicals.

Among the halogenated meta-aminophenols of formula (I), mention may be made more particularly of 3-amino-6-chlorophenol, 3-amino-6-bromophenol, 3-( $\beta$ -aminoethyl)amino-6-chlorophenol, 3-( $\beta$ -hydroxyethyl)-amino-6-chlorophenol and 3-amino-2-chloro-6-methylphenol, and the addition salts thereof with an acid.

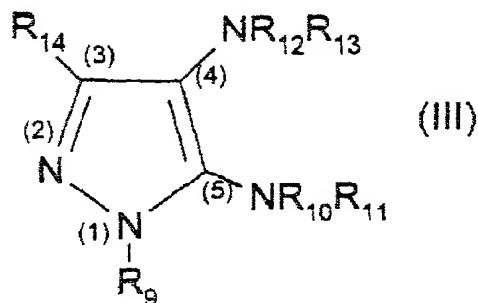
Among the diaminopyrazoles which can be used as oxidation bases in the dye compositions in accordance with the invention, mention may be made more particularly of:

a) the diaminopyrazoles of formula (II) below, and the addition salts thereof with an acid:



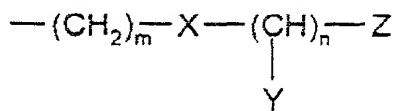
in which:

- $R_5$  represents a hydrogen atom, a  $C_1$ - $C_6$  alkyl radical, a  $C_2$ - $C_4$  hydroxyalkyl radical, a benzyl radical, a phenyl radical, a benzyl radical substituted with a halogen atom or with a  $C_1$ - $C_4$  alkyl or  $C_1$ - $C_4$  alkoxy group, or forms, with the nitrogen atom of the group  $NR_7R_8$  in position 5, a hexahydropyridazine or tetrahydropyrazole heterocycle which is optionally monosubstituted with a  $C_1$ - $C_4$  alkyl group;
  - $R_6$  and  $R_7$  which may be identical or different, represent a hydrogen atom, a  $C_1$ - $C_4$  alkyl radical, a  $C_2$ - $C_4$  hydroxyalkyl radical, a benzyl radical or a phenyl radical;
  - $R_8$  represents a hydrogen atom, or a  $C_1$ - $C_6$  alkyl or  $C_2$ - $C_4$  hydroxyalkyl radical; with the proviso that  $R_6$  represents a hydrogen atom when  $R_5$  represents a substituted benzyl radical or forms a heterocycle with the nitrogen atom of the group  $NR_7R_8$  in position 5;
- b) the diaminopyrazoles of formula (III) below, and the addition salts thereof with an acid:



in which:

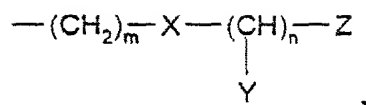
- $R_9$ ,  $R_{10}$ ,  $R_{11}$ ,  $R_{12}$  and  $R_{13}$ , which may be identical or different, represent a hydrogen atom; a linear or branched  $C_1$ - $C_6$  alkyl radical; a  $C_2$ - $C_4$  hydroxyalkyl radical; a  $C_2$ - $C_4$  aminoalkyl radical; a phenyl radical; a phenyl radical substituted with a halogen atom or a  $C_1$ - $C_4$  alkyl,  $C_1$ - $C_4$  alkoxy, nitro, trifluoromethyl, amino or  $C_1$ - $C_4$  alkylamino radical; a benzyl radical; a benzyl radical substituted with a halogen atom or with a  $C_1$ - $C_4$  alkyl,  $C_1$ - $C_4$  alkoxy, methylenedioxy or amino radical; or a radical



- in which  $m$  and  $n$  are integers, which may be identical or different, between 1 and 3 inclusive,  $X$  represents an oxygen atom or an  $NH$  group,  $Y$  represents a hydrogen atom or a methyl radical, and  $Z$  represents a methyl radical, a group  $OR$  or  $NRR'$  in which  $R$  and  $R'$ , which may be identical or different, denote a hydrogen atom, a methyl radical or an ethyl radical,

it being understood that when  $R_{10}$  represents a hydrogen atom, then  $R_{11}$  can also represent an amino or  $C_1$ - $C_4$  alkylamino radical,

- $R_{14}$  represents a linear or branched  $C_1$ - $C_6$  alkyl radical; a  $C_1$ - $C_4$  hydroxyalkyl radical; a  $C_1$ - $C_4$  aminoalkyl radical; a  $(C_1-C_4)$ alkylamino $(C_1-C_4)$ alkyl radical; a di $(C_1-C_4)$ alkylamino $(C_1-C_4)$ alkyl radical; a hydroxy $(C_1-C_4)$ alkylamino $(C_1-C_4)$ alkyl radical; a  $(C_1-C_4)$ alkoxymethyl radical; a phenyl radical; a phenyl radical substituted with a halogen atom or with a  $C_1$ - $C_4$  alkyl,  $C_1$ - $C_4$  alkoxy, nitro, trifluoromethyl, amino or  $C_1$ - $C_4$  alkylamino radical; a benzyl radical; a benzyl radical substituted with a halogen atom or with a  $C_1$ - $C_4$  alkyl,  $C_1$ - $C_4$  alkoxy, nitro, trifluoromethyl, amino or  $C_1$ - $C_4$  alkylamino radical; a heterocycle chosen from thiophene, furan and pyridine, or alternatively a radical  $-(CH_2)_p-O-(CH_2)_q-$  OR", in which p and q are integers, which may be identical or different, between 1 and 3 inclusive, and R" represents a hydrogen atom or a methyl radical,
- it being understood that, in formula (III) above,
- at least one of the radicals  $R_{10}$ ,  $R_{11}$ ,  $R_{12}$  and  $R_{13}$  represents a hydrogen atom,
  - when  $R_{10}$ , or  $R_{12}$ , respectively, represents a substituted or unsubstituted phenyl radical, or a benzyl radical or a radical



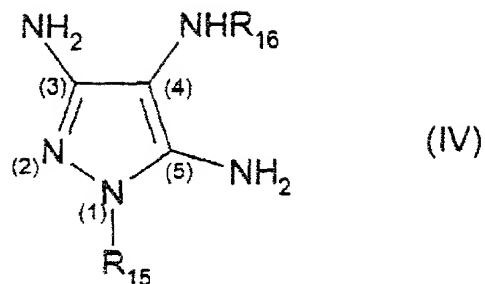
then R<sub>11</sub>, or R<sub>13</sub>, respectively, cannot represent any of these three radicals,

- 5 - when R<sub>12</sub> and R<sub>13</sub> simultaneously represent a hydrogen atom, then R<sub>9</sub> can form, with R<sub>10</sub> and R<sub>11</sub>, a hexahydropyrimidine or tetrahydroimidazole heterocycle which is optionally substituted with a C<sub>1</sub>-C<sub>4</sub> alkyl or 1,2,4-tetrazole radical,
- 10 - when R<sub>10</sub>, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub> represent a hydrogen atom or a C<sub>1</sub>-C<sub>6</sub> alkyl radical, then R<sub>9</sub> or R<sub>14</sub> can also represent a 2-, 3- or 4-pyridyl, 2- or 3-thienyl or 2- or 3-furyl heterocyclic residue which is optionally substituted with a methyl radical or alternatively a cyclohexyl
- 15 radical.

Among the triaminopyrazoles which can be used as oxidation bases in the dye compositions in accordance with the invention, mention may be made more particularly of the compounds of formula (IV) below,

20 and the addition salts thereof with an acid:





in which:

- $R_{15}$  and  $R_{16}$ , which may be identical or different, represent a hydrogen atom or a  $C_1$ - $C_4$  alkyl or  $C_2$ - $C_4$  hydroxyalkyl radical.

Among the diaminopyrazoles of formula (II) above, mention may be made more particularly of 4,5-diamino-1-(4'-methoxybenzyl)pyrazole, 4,5-diamino-1-(4'-methylbenzyl)pyrazole, 4,5-diamino-1-(4'-chlorobenzyl)pyrazole, 4,5-diamino-1-(3'-methoxybenzyl)pyrazole, 4-amino-1-(4'-methoxybenzyl)-5-methylaminopyrazole, 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(4'-methoxybenzyl)pyrazole, 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-methylpyrazole, 4-amino-(3)5-methylaminopyrazole, 3-(5)4-diaminopyrazole, 4,5-diamino-1-methylpyrazole, 4,5-diamino-1-benzylpyrazole, 3-amino-4,5,7,8-tetrahydropyrazolo[1,5-a]pyrimidine, 7-amino-2,3-dihydro-1H-imidazolo[1,2-b]pyrazole and 3-amino-8-methyl-4,5,7,8-tetrahydropyrazolo[1,5-a]pyrimidine, and the addition salts thereof with an acid.

The diaminopyrazoles of formula (III) are known compounds which can be prepared according to the synthetic process as described, for example, in French patent application FR-A-2 733 749.

- 5           Among the diaminopyrazoles of formula (III) above, mention may be made more particularly of:
- 1-benzyl-4,5-diamino-3-methylpyrazole,
  - 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(4'-methoxyphenyl)-pyrazole,
  - 10 - 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(4'-methylphenyl)-pyrazole,
  - 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(3'-methylphenyl)-pyrazole,
  - 4,5-diamino-3-methyl-1-isopropylpyrazole,
  - 15 - 4,5-diamino-3-(4'-methoxyphenyl)-1-isopropylpyrazole,
  - 4,5-diamino-1-ethyl-3-methylpyrazole,
  - 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-methylpyrazole,
  - 4,5-diamino-1-ethyl-3-hydroxymethylpyrazole,
  - 20 - 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-tert-butylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-phenylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-(2'-methoxyphenyl)-pyrazole,
  - 25 - 4,5-diamino-3-hydroxymethyl-1-(3'-methoxyphenyl)-pyrazole,

- 4,5-diamino-3-hydroxymethyl-1-(4'-methoxyphenyl)-pyrazole,
- 1-benzyl-4,5-diamino-3-hydroxymethylpyrazole,
- 4,5-diamino-3-methyl-1-(2'-methoxyphenyl)pyrazole,
- 5 - 4,5-diamino-3-methyl-1-(3'-methoxyphenyl)pyrazole,
- 4,5-diamino-3-methyl-1-(4'-methoxyphenyl)pyrazole,
- 3-aminomethyl-4,5-diamino-1-methylpyrazole,
- 3-aminomethyl-4,5-diamino-1-ethylpyrazole,
- 3-aminomethyl-4,5-diamino-1-isopropylpyrazole,
- 10 - 3-aminomethyl-4,5-diamino-1-tert-butylpyrazole,
- 4,5-diamino-3-dimethylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-dimethylaminomethyl-1-ethylpyrazole,
- 4,5-diamino-3-dimethylaminomethyl-1-isopropylpyrazole,
- 15 - 4,5-diamino-3-dimethylaminomethyl-1-tert-butylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-ethylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-isopropylpyrazole,
- 20 - 4,5-diamino-3-ethylaminomethyl-1-tert-butylpyrazole,
- 4,5-diamino-3-methylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-methylaminomethyl-1-isopropylpyrazole,
- 4,5-diamino-1-ethyl-3-methylaminomethylpyrazole,
- 1-tert-butyl-4,5-diamino-3-methylaminomethylpyrazole,
- 25 - 4,5-diamino-3-[( $\beta$ -hydroxyethyl)aminomethyl]-1-methylpyrazole,

- 4,5-diamino-3-[( $\beta$ -hydroxyethyl)aminomethyl]-1-isopropylpyrazole,
- 4,5-diamino-1-ethyl-3-[( $\beta$ -hydroxyethyl)aminomethyl]-pyrazole,
- 5 - 1-tert-butyl-4,5-diamino-3-[( $\beta$ -hydroxyethyl)-aminomethyl]pyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1,3-dimethylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-isopropyl-3-methylpyrazole,
- 10 - 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-ethyl-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-tert-butyl-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-phenyl-3-methylpyrazole,
- 15 - 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(2-methoxyphenyl)-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(3-methoxyphenyl)-3-methylpyrazole,
- 20 - 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(4-methoxyphenyl)-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-benzyl-3-methylpyrazole,
- 4-amino-1-ethyl-3-methyl-5-methylaminopyrazole,

- 4-amino-1-tert-butyl-3-methyl-5-methylaminopyrazole,  
- 4,5-diamino-1,3-dimethylpyrazole,  
- 4,5-diamino-3-tert-butyl-1-methylpyrazole,  
- 4,5-diamino-1-tert-butyl-3-methylpyrazole,  
5 - 4,5-diamino-1-methyl-3-phenylpyrazole,  
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-methylpyrazole,  
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-phenylpyrazole,  
- 4,5-diamino-1-methyl-3-(2'-chlorophenyl)pyrazole,  
- 4,5-diamino-1-methyl-3-(4'-chlorophenyl)pyrazole,  
10 - 4,5-diamino-1-methyl-3-(3'-trifluoromethylphenyl)-  
pyrazole,  
- 4,5-diamino-1,3-diphenylpyrazole,  
- 4,5-diamino-3-methyl-1-phenylpyrazole,  
- 4-amino-1,3-dimethyl-5-phenylaminopyrazole,  
15 - 4-amino-1-ethyl-3-methyl-5-phenylaminopyrazole,  
- 4-amino-1,3-dimethyl-5-methylaminopyrazole,  
- 4-amino-3-methyl-1-isopropyl-5-methylaminopyrazole,  
- 4-amino-3-isobutoxymethyl-1-methyl-5-methylamino-  
pyrazole,  
20 - 4-amino-3-methoxyethoxymethyl-1-methyl-5-methylamino-  
pyrazole,  
- 4-amino-3-hydroxymethyl-1-methyl-5-methylamino-  
pyrazole,  
- 4-amino-1,3-diphenyl-5-phenylaminopyrazole,  
25 - 4-amino-3-methyl-5-methylamino-1-phenylpyrazole,

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- 4-amino-1,3-dimethyl-5-hydrazinopyrazole,
  - 5-amino-3-methyl-4-methylamino-1-phenylpyrazole,
  - 5-amino-1-methyl-4-(N,N-methylphenyl)amino-3-(4'-chlorophenyl)pyrazole,
  - 5 - 5-amino-3-ethyl-1-methyl-4-(N,N-methylphenyl)amino-pyrazole,
  - 5-amino-1-methyl-4-(N,N-methylphenyl)amino-3-phenylpyrazole,
  - 5-amino-3-ethyl-4-(N,N-methylphenyl)aminopyrazole,
  - 10 - 5-amino-4-(N,N-methylphenyl)amino-3-phenylpyrazole,
  - 5-amino-4-(N,N-methylphenyl)amino-3-(4'-methylphenyl)pyrazole,
  - 5-amino-3-(4'-chlorophenyl)-4-(N,N-methylphenyl)-aminopyrazole,
  - 15 - 5-amino-3-(4'-methoxyphenyl)-4-(N,N-methylphenyl)-aminopyrazole,
  - 4-amino-5-methylamino-3-phenylpyrazole,
  - 4-amino-5-ethylamino-3-phenylpyrazole,
  - 4-amino-5-ethylamino-3-(4'-methylphenyl)pyrazole,
  - 20 - 4-amino-3-phenyl-5-propylaminopyrazole,
  - 4-amino-5-butylamino-3-phenylpyrazole,
  - 4-amino-3-phenyl-5-phenylaminopyrazole,
  - 4-amino-5-benzylamino-3-phenylpyrazole,
  - 4-amino-5-(4'-chlorophenyl)amino-3-phenylpyrazole,
  - 25 - 4-amino-3-(4'-chlorophenyl)-5-phenylaminopyrazole,
  - 4-amino-3-(4'-methoxyphenyl)-5-phenylaminopyrazole,

- 5 and the addition salts thereof with an acid.

- 4,5-diamino-1,3-dimethylpyrazole,
- 4,5-diamino-3-methyl-1-phenylpyrazole,

- Among the triaminopyrazoles of formula (IV)

25 above, mention may be made more particularly of 3,4,5-  
triaminopyrazole, 1-methyl-3,4,5-triaminopyrazole, 3,5-

diamino-1-methyl-4-methylaminopyrazole and 3,5-diamino-4-( $\beta$ -hydroxyethyl)amino-1-methylpyrazole, and the addition salts thereof with an acid.

The diaminopyrazole(s) and/or the triaminopyrazole(s) in accordance with the invention and/or the corresponding addition salt(s) with an acid preferably represent(s) from 0.0005 to 12% by weight approximately relative to the total weight of the dye composition, and even more preferably from 0.005 to 6% by weight approximately relative to this weight.

The halogenated meta-aminophenol(s) of formula (I) in accordance with the invention and/or the corresponding addition salt(s) with an acid preferably represent(s) from 0.0001 to 5% by weight approximately 15 relative to the total weight of the dye composition, and even more preferably from 0.005 to 3% by weight approximately relative to this weight.

The dye compositions in accordance with the invention can contain other couplers conventionally used for oxidation dyeing, other than the halogenated meta-aminophenols of formula (I), and/or other oxidation bases conventionally used for oxidation dyeing, other than a diaminopyrazole and a triaminopyrazole and/or direct dyes, in particular in order to modify the shades or to enrich them with glints.



In general, the addition salts with an acid which can be used in the context of the dye compositions of the invention (oxidation bases and couplers) are chosen in particular from the  
5 hydrochlorides, hydrobromides, sulphates, tartrates, lactates and acetates.

The medium which is suitable for dyeing (or support) generally consists of water or of a mixture of water and at least one organic solvent to dissolve the  
10 compounds which would not be sufficiently soluble in water. Organic solvents which may be mentioned, for example, are C<sub>1</sub>-C<sub>4</sub> lower alkanols, such as ethanol and isopropanol; glycerol; glycols and glycol ethers such as 2-butoxyethanol, propylene glycol, propylene glycol  
15 monomethyl ether, diethylene glycol monoethyl ether and monomethyl ether, as well as aromatic alcohols such as benzyl alcohol or phenoxyethanol, similar products and mixtures thereof.

The solvents can be present in proportions  
20 preferably of between 1 and 40% by weight approximately relative to the total weight of the dye composition, and even more preferably between 5 and 30% by weight approximately.

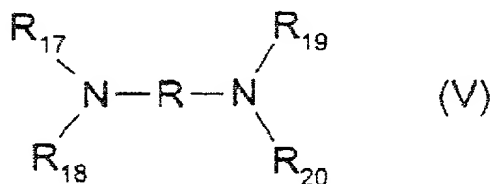
The pH of the dye composition in accordance  
25 with the invention is generally between 3 and 12 approximately and even more preferably between 5 and 11

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approximately. It can be adjusted to the desired value by means of acidifying or basifying agents usually used in the dyeing of keratin fibres.

Among the acidifying agents which may be mentioned, for example, are inorganic or organic acids such as hydrochloric acid, orthophosphoric acid, carboxylic acids such as tartaric acid, citric acid and lactic acid, and sulphonic acids.

Among the basifying agents which may be mentioned, for example, are aqueous ammonia, alkaline carbonates, alkanolamines such as mono-, di- and triethanolamine and derivatives thereof, sodium hydroxide, potassium hydroxide and the compounds of formula (V) below:



in which R is a propylene residue optionally substituted with a hydroxyl group or a C<sub>1</sub>-C<sub>4</sub> alkyl radical; R<sub>17</sub>, R<sub>18</sub>, R<sub>19</sub> and R<sub>20</sub>, which may be identical or different, represent a hydrogen atom or a C<sub>1</sub>-C<sub>4</sub> alkyl or C<sub>1</sub>-C<sub>4</sub> hydroxyalkyl radical.

The dye composition according to the invention can also contain various adjuvants conventionally used in compositions for dyeing the

hair, such as anionic, cationic, nonionic, amphoteric  
or zwitterionic surfactants or mixtures thereof,  
anionic, cationic, nonionic, amphoteric or zwitterionic  
polymers or mixtures thereof, inorganic or organic  
5 thickeners, antioxidants, penetrating agents,  
sequestering agents, fragrances, buffers, dispersants,  
conditioners such as, for example, volatile or non-  
volatile, modified or unmodified silicones, film-  
forming agents, ceramides, preserving agents and  
10 opacifiers.

Needless to say, a person skilled in the art  
will take care to select this or these optional  
additional compound(s) such that the advantageous  
properties intrinsically associated with the  
15 combination in accordance with the invention are not,  
or are not substantially, adversely affected by the  
addition(s) envisaged.

The dye composition according to the  
invention can be in various forms, such as in the form  
20 of liquids, creams or gels or in any other form which  
is suitable for dyeing keratin fibres, and in  
particular human hair.

A subject of the invention is also a process  
for dyeing keratin fibres, and in particular human  
25 keratin fibres such as the hair, using the dye  
composition as defined above.

According to this process, the dye composition as defined above is applied to the fibres, the colour being developed at acidic, neutral or alkaline pH with the aid of an oxidizing agent which is added to the dye composition just at the time of use, or which is present in an oxidizing composition that is applied simultaneously or sequentially.

According to one particularly preferred embodiment of the dyeing process according to the invention, the dye composition described above is mixed, at the time of use, with an oxidizing composition containing, in a medium which is suitable for dyeing, at least one oxidizing agent present in an amount which is sufficient to develop a coloration. The mixture obtained is then applied to the keratin fibres and is left to stand on them for 3 to 60 minutes approximately, preferably 5 to 40 minutes approximately, after which the fibres are rinsed, washed with shampoo, rinsed again and dried.

The oxidizing agent present in the oxidizing composition as defined above can be chosen from the oxidizing agents conventionally used for the oxidation dyeing of keratin fibres, and among which mention may be made of hydrogen peroxide, urea peroxide, alkali metal bromates, persalts such as perborates,

percarbonates and persulphates, and peracids. Hydrogen peroxide is particularly preferred.

The pH of the oxidizing composition containing the oxidizing agent as defined above is such that after mixing with the dye composition, the pH of the resulting composition applied to the keratin fibres preferably ranges between 3 and 12 approximately and even more preferably between 5 and 11. It is adjusted to the desired value by means of acidifying or basifying agents usually used in the dyeing of keratin fibres and as defined above.

The oxidizing composition as defined above can also contain various adjuvants conventionally used in compositions for dyeing the hair and as defined above.

The composition which is finally applied to the keratin fibres can be in various forms, such as in the form of liquids, creams or gels or in any other form which is suitable for dyeing keratin fibres, and in particular human hair.

Another subject of the invention is a multi-compartment dyeing device or "kit" or any other multi-compartment packaging system, a first compartment of which contains the dye composition as defined above, and a second compartment of which contains the oxidizing composition as defined above. These devices

can be equipped with a means for applying the desired mixture to the hair, such as the devices described in patent FR-2 586 913 in the name of the Applicant.

The examples which follow are intended to  
5 illustrate the invention without, however, limiting its scope.

### EXAMPLES

#### COMPARATIVE DYEING EXAMPLES 1 TO 4

The dye compositions below, in accordance  
10 with the invention, were prepared (contents in grams):

| EXAMPLE   | 1 (*) | 2     | 3     | 4     |
|---|-------|-------|-------|-------|
| 4,5-diamino-1-ethyl-3-methylpyrazole dihydrochloride (oxidation base)                   | 0.639 | 0.639 | 0.639 | 0.639 |
| 3-aminophenol (coupler not forming part of the invention)                               | 0.327 | -     | -     | -     |
| 3-amino-6-chlorophenol (coupler in accordance with the invention)                       | -     | 0.431 | -     | -     |
| 3-( $\beta$ -aminoethyl)amino-6-chlorophenol (coupler in accordance with the invention) | -     | -     | 0.560 | -     |

(\*) : example not forming part of the invention

|   |             |
|---|-------------|
| - Oleyl alcohol polyglycerated with<br>2 mol of glycerol  | 4.0 g       |
| - Oleyl alcohol polyglycerated with<br>4 mol of glycerol, containing 78% active<br>material (A.M.)                | 5.69 g A.M. |
| - Oleic acid  | 3.0 g       |
| - Oleylamine containing 2 mol of<br>ethylene oxide, sold under the tradename<br>Ethomeen O12® by the company Akzo | 7.0 g       |
| - Diethylaminopropyl laurylamino<br>succinamate, sodium salt, containing<br>55% A.M.                              | 3.0 g A.M.  |
| - Oleyl alcohol   | 5.0 g       |
| - Oleic acid diethanolamide   | 12.0 g      |
| - Propylene glycol  | 3.5 g       |
| - Ethyl alcohol   | 7.0 g       |

|  |              |
|--|--------------|
| - Dipropylene glycol   | 0.5 g        |
| - Propylene glycol monomethyl ether                                | 9.0 g        |
| - Sodium metabisulphite as an aqueous solution containing 35% A.M. | 0.455 g A.M. |
| - Ammonium acetate   | 0.8 g        |
| - Antioxidant, sequestering agent                                  | q.s.         |
| - Fragrance, preserving agent                                      | q.s.         |
| - Aqueous ammonia containing 20% $\text{NH}_3$                     | 10 g         |

It is important to note that each of the dye compositions 1 to 4 above contains the same molar amount of coupler, i.e.  $3 \times 10^{-3}$  mol.

5           At the time of use, each dye composition above was mixed with an equal amount by weight of an oxidizing composition consisting of a 20-volumes aqueous hydrogen peroxide solution (6% by weight).

Each resulting composition was applied for 30  
10 minutes to locks of natural grey hair containing 90% white hairs. The locks of hair were then rinsed, washed with a standard shampoo and then dried.

The locks of dyed hair were then subjected to a test of resistance to the action of perspiration.

15           The colour of the locks of hair dyed with compositions 1 to 4 was evaluated in the Munsell system using a Minolta® CM 2002 colorimeter, before the test of resistance to the action of perspiration.



According to the Munsell notation, a colour is defined by the expression  $H\ V\ /\ C$  in which the three parameters respectively denote the shade or Hue (H), the intensity or Value (V) and the purity or Chromaticity (C), the oblique line in this expression simply being a convention and not indicating a ratio.

The locks of dyed hair were then subjected to the test of resistance to the action of perspiration.

To do this, the locks of dyed hair were immersed in a crystallizing dish covered with a watch glass and containing a solution of synthetic sweat of the following composition:

|                                |         |
|--------------------------------|---------|
| - NaCl                         | 1.0 g   |
| - Potassium hydrogen phosphate | 0.1 g   |
| 15 - Histidine                 | 0.025 g |
| - Lactic acid qs               | pH 3.2  |
| - Distilled water qs           | 100 g   |

The locks of dyed hair were left to stand in this synthetic sweat solution for 48 hours at 37°C. The locks were then rinsed, followed by drying.

The colour of the locks was then re-evaluated in the Munsell system using a Minolta® CM 2002 colorimeter.

25 The difference between the colour of the lock before the test of resistance to perspiration and the

colour of the lock after the test of resistance to perspiration was calculated by applying the Nickerson formula

$$\Delta E = 0.4C_0dH+6dV+3dC$$

5 as described, for example, in "Couleur, Industrie et Technique [Colour, Industry and Technique]"; pages 14-17; vol. No 5; 1978.

In this formula,  $\Delta E$  represents the difference in colour between two locks,  $\Delta H$ ,  $\Delta V$  and  $\Delta C$  represent  
10 the variation in absolute value of the parameters H, V and C, and  $C_0$  represents the purity of the lock relative to which it is desired to evaluate the colour difference.

The degradation of the colour is  
15 proportionately greater the larger the value of  $\Delta E$ .

The results are given in the table below:

| EXAMPLE | Colour of<br>the hair<br>before the<br>test | Colour of<br>the hair<br>after the<br>test | Degradation of the<br>colour |            |            |            |
|---------|---|--|------------------------------|------------|------------|------------|
|         |   |  | $\Delta H$                   | $\Delta V$ | $\Delta C$ | $\Delta E$ |
| 1(*)    | 9.2 RP<br>3/3/2.6                           | 2.5 YR<br>3.9/2.1                          | 13.3                         | 0.6        | 0.5        | 18.9       |
| 2       | 2.6 RP<br>3.0/3.7                           | 6.1 RP<br>3.5/3.2                          | 3.5                          | 0.5        | 0.5        | 9.7        |
| 3       | 2.6 RP<br>3.0/2.9                           | 4.1 RP<br>3.0/2.9                          | 1.5                          | 0          | 0          | 1.7        |
| 4       | 2.7 RP<br>3.2/3.2                           | 6.1 RP<br>3.5/3.0                          | 3.4                          | 0.3        | 0.2        | 6.8        |

These results show that the coloration obtained using the dye composition of Example 1 not forming part of the invention, since it contains a combination of a diaminopyrazole and a non-halogenated meta-aminophenol, is markedly less resistant to the action of perspiration than the colorations obtained using the compositions of Examples 2 to 4, all of which form part of the invention since they contain a combination of a diaminopyrazole and a meta-aminophenol which is halogenated ortho to the phenol.

DYEING EXAMPLES 5 TO 8

The dye compositions below, in accordance with the invention, were prepared (contents in grams):

| EXAMPLE   | 5     | 6     | 7     | 8     |
|---|-------|-------|-------|-------|
| 4,5-diaminopyrazole dihydrochloride (oxidation base)                                    | 0.513 | -     | -     | -     |
| 1-methyl-4,5-diaminopyrazole dihydrochloride (oxidation base)                           | -     | 0.555 | 0.555 | 0.555 |
| 3-amino-2-chloro-6-methylphenol (coupler in accordance with the invention)              | 0.473 | 0.473 | -     | -     |
| 3-amino-6-chlorophenol (coupler in accordance with the invention)                       | -     | -     | 0.431 | -     |
| 3-( $\beta$ -aminoethyl)amino-6-chlorophenol (coupler in accordance with the invention) | -     | -     | -     | 0.560 |
| Common dye support  | (**)  | (**)  | (**)  | (**)  |
| Demineralized water qs  | 100 g | 100 g | 100 g | 100 g |

5 (\*\*) common dye support:

This is identical to the one used for Examples 1 to 4 above.

At the time of use, each dye composition 5 above was mixed with an equal amount by weight of an oxidizing composition consisting of a 20-volumes aqueous hydrogen peroxide solution (6% by weight).

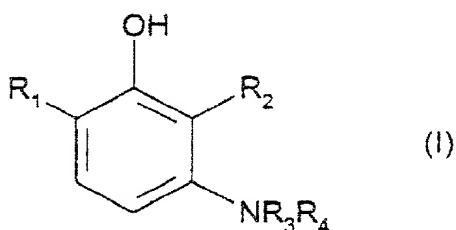
Each resulting composition was applied for 30 minutes to locks of natural grey hair containing 90% 10 white hairs. The locks of hair were then rinsed, washed with a standard shampoo and then dried.

The locks were dyed in the shades given in the table below:

| EXAMPLE | SHADE OBTAINED |
|---------|----------------|
| 5       | Red-coppery    |
| 6       | Red-coppery    |
| 7       | Red-iridescent |
| 8       | Iridescent red |

## CLAIMS

1. Composition for the oxidation dyeing of human keratin fibres and in particular human keratin fibres such as the hair, characterized in that it comprises, in a medium which is suitable for dyeing:
- at least one oxidation base chosen from diaminopyrazoles and triaminopyrazoles;
  - and at least one coupler chosen from the halogenated meta-aminophenols of formula (I) below, and the addition salts thereof with an acid:



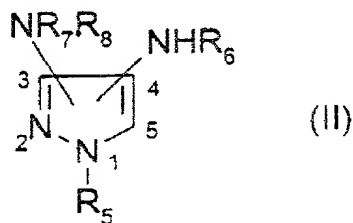
in which:

- $R_1$  and  $R_2$ , which may be identical or different, represent a hydrogen atom, a halogen atom such as chlorine, bromine, iodine or fluorine, a  $C_1$ - $C_4$  alkyl radical, a  $C_1$ - $C_4$  monohydroxyalkyl radical, a  $C_2$ - $C_4$  polyhydroxyalkyl radical, a  $C_1$ - $C_4$  alkoxy radical, a  $C_1$ - $C_4$  monohydroxyalkoxy radical or a  $C_2$ - $C_4$  polyhydroxyalkoxy radical;
- $R_3$  and  $R_4$ , which may be identical or different, represent a hydrogen atom, a  $C_1$ - $C_4$  alkyl radical, a  $C_1$ - $C_4$  monohydroxyalkyl radical, a  $C_2$ - $C_4$  polyhydroxyalkyl radical or a  $C_1$ - $C_4$  monoaminoalkyl radical;

it being understood that at least one of the radicals  $R_1$  and  $R_2$  represents a halogen atom.

2. Composition according to Claim 1, characterized in that the halogenated meta-aminophenols of formula (I) are chosen from 3-amino-6-chlorophenol, 3-amino-6-bromophenol, 3-( $\beta$ -aminoethyl)amino-6-chlorophenol, 3-( $\beta$ -hydroxyethyl)amino-6-chlorophenol and 3-amino-2-chloro-6-methylphenol, and the addition salts thereof with an acid.

3. Composition according to Claim 1 or 2, characterized in that the diaminopyrazoles which can be used as oxidation bases are chosen from:  
a) the diaminopyrazoles of formula (II) below, and the addition salts thereof with an acid:



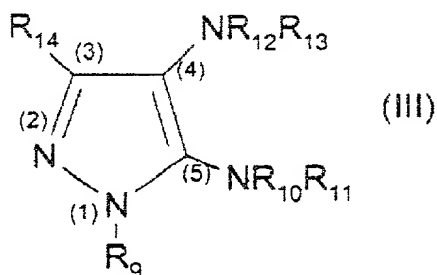
in which:

-  $R_5$  represents a hydrogen atom, a  $C_1$ - $C_6$  alkyl radical, a  $C_2$ - $C_4$  hydroxyalkyl radical, a benzyl radical, a phenyl radical, a benzyl radical substituted with a halogen atom or with a  $C_1$ - $C_4$  alkyl or  $C_1$ - $C_4$  alkoxy group, or forms, with the nitrogen atom of the group  $NR_7R_8$  in position 5, a hexahydropyridazine or tetrahydropyrazole

heterocycle which is optionally monosubstituted with a C<sub>1</sub>-C<sub>4</sub> alkyl group;

- R<sub>6</sub> and R<sub>7</sub> which may be identical or different, represent a hydrogen atom, a C<sub>1</sub>-C<sub>4</sub> alkyl radical, a C<sub>2</sub>-C<sub>4</sub> hydroxyalkyl radical, a benzyl radical or a phenyl radical;

- R<sub>8</sub> represents a hydrogen atom, or a C<sub>1</sub>-C<sub>6</sub> alkyl or C<sub>2</sub>-C<sub>4</sub> hydroxyalkyl radical; with the proviso that R<sub>6</sub> represents a hydrogen atom when R<sub>5</sub> represents a substituted benzyl radical or forms a heterocycle with the nitrogen atom of the group NR<sub>7</sub>R<sub>8</sub> in position 5;  
b) the diaminopyrazoles of formula (III) below, and the addition salts thereof with an acid:

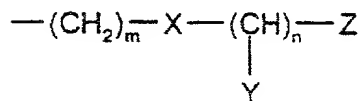


in which:

- R<sub>9</sub>, R<sub>10</sub>, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub>, which may be identical or different, represent a hydrogen atom; a linear or branched C<sub>1</sub>-C<sub>6</sub> alkyl radical; a C<sub>2</sub>-C<sub>4</sub> hydroxyalkyl radical; a C<sub>2</sub>-C<sub>4</sub> aminoalkyl radical; a phenyl radical; a phenyl radical substituted with a halogen atom or a C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy, nitro, trifluoromethyl, amino or C<sub>1</sub>-C<sub>4</sub> alkylamino radical; a benzyl radical; a benzyl



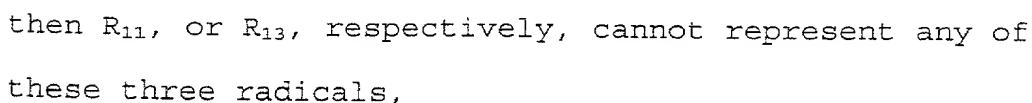
radical substituted with a halogen atom or with a C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy, methylenedioxy or amino radical; or a radical



- 5 in which m and n are integers, which may be identical or different, between 1 and 3 inclusive, X represents an oxygen atom or an NH group, Y represents a hydrogen atom or a methyl radical, and Z represents a methyl radical, a group OR or NRR' in which R and R', which
- 10 may be identical or different, denote a hydrogen atom, a methyl radical or an ethyl radical, it being understood that when R<sub>10</sub> represents a hydrogen atom, then R<sub>11</sub> can also represent an amino or C<sub>1</sub>-C<sub>4</sub> alkylamino radical,
- 15 - R<sub>14</sub> represents a linear or branched C<sub>1</sub>-C<sub>6</sub> alkyl radical; a C<sub>1</sub>-C<sub>4</sub> hydroxyalkyl radical; a C<sub>1</sub>-C<sub>4</sub> aminoalkyl radical; a (C<sub>1</sub>-C<sub>4</sub>)alkylamino(C<sub>1</sub>-C<sub>4</sub>)alkyl radical; a di(C<sub>1</sub>-C<sub>4</sub>)alkylamino(C<sub>1</sub>-C<sub>4</sub>)alkyl radical; a hydroxy(C<sub>1</sub>-C<sub>4</sub>)alkylamino(C<sub>1</sub>-C<sub>4</sub>)alkyl radical; a (C<sub>1</sub>-C<sub>4</sub>)alkoxymethyl
- 20 radical; a phenyl radical; a phenyl radical substituted with a halogen atom or with a C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy, nitro, trifluoromethyl, amino or C<sub>1</sub>-C<sub>4</sub> alkylamino radical; a benzyl radical; a benzyl radical substituted with a halogen atom or with a C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy,
- 25 nitro, trifluoromethyl, amino or C<sub>1</sub>-C<sub>4</sub> alkylamino

5 "R" represents a hydrogen atom or a methyl radical,  
it being understood that, in formula (III) above,  
- at least one of the radicals  $R_{10}$ ,  $R_{11}$ ,  $R_{12}$  and  $R_{13}$   
represents a hydrogen atom,

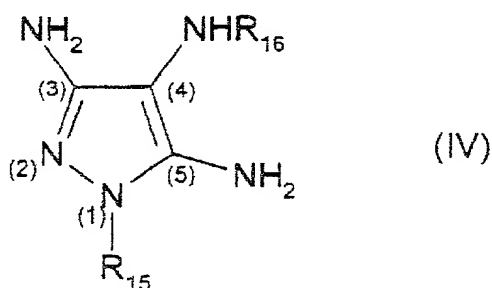
10 substituted or unsubstituted phenyl radical, or a  
benzyl radical or a radical



20 - when R<sub>10</sub>, R<sub>11</sub>, R<sub>12</sub> and R<sub>13</sub> represent a hydrogen atom or a C<sub>1</sub>-C<sub>6</sub> alkyl radical, then R<sub>9</sub> or R<sub>14</sub> can also represent a 2-, 3- or 4-pyridyl, 2- or 3-thienyl or 2- or 3-furyl heterocyclic residue which is optionally substituted

with a methyl radical or alternatively a cyclohexyl radical.

4. Composition according to Claim 1 or 2, characterized in that the triaminopyrazoles which can be used as oxidation bases are chosen from the compounds of formula (IV) below, and the addition salts thereof with an acid:



in which:

- 10 -  $R_{15}$  and  $R_{16}$ , which may be identical or different, represent a hydrogen atom or a  $C_1$ - $C_4$  alkyl or  $C_2$ - $C_4$  hydroxyalkyl radical.

5. Composition according to Claim 3, characterized in that the diaminopyrazoles of formula (II) are chosen from 4,5-diamino-1-(4'-methoxybenzyl)-pyrazole, 4,5-diamino-1-(4'-methylbenzyl)pyrazole, 4,5-diamino-1-(4'-chlorobenzyl)pyrazole, 4,5-diamino-1-(3'-methoxybenzyl)pyrazole, 4-amino-1-(4'-methoxybenzyl)-5-methylaminopyrazole, 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(4'-methoxybenzyl)pyrazole, 4-amino-5-( $\beta$ -hydroxyethyl)-amino-1-methylpyrazole, 4-amino-(3)5-methylamino-

5 methyl-4,5,7,8-tetrahydropyrazolo[1,5-a]pyrimidine, and  
the addition salts thereof with an acid.

6. Composition according to Claim 3,  
characterized in that the diaminopyrazoles of formula  
(III) are chosen from:

- 10 - 1-benzyl-4,5-diamino-3-methylpyrazole,  
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(4'-methoxyphenyl)-  
pyrazole,  
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(4'-methylphenyl)-  
pyrazole,  
15 - 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-(3'-methylphenyl)-  
pyrazole,  
- 4,5-diamino-3-methyl-1-isopropylpyrazole,  
- 4,5-diamino-3-(4'-methoxyphenyl)-1-isopropylpyrazole,  
- 4,5-diamino-1-ethyl-3-methylpyrazole,  
20 - 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole,  
- 4,5-diamino-3-hydroxymethyl-1-methylpyrazole,  
- 4,5-diamino-1-ethyl-3-hydroxymethylpyrazole,  
- 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole,  
- 4,5-diamino-3-hydroxymethyl-1-tert-butylpyrazole,  
25 - 4,5-diamino-3-hydroxymethyl-1-phenylpyrazole,

- 4,5-diamino-3-hydroxymethyl-1-(2'-methoxyphenyl)-pyrazole,
- 4,5-diamino-3-hydroxymethyl-1-(3'-methoxyphenyl)-pyrazole,
- 5 - 4,5-diamino-3-hydroxymethyl-1-(4'-methoxyphenyl)-pyrazole,
- 1-benzyl-4,5-diamino-3-hydroxymethylpyrazole,
- 4,5-diamino-3-methyl-1-(2'-methoxyphenyl)pyrazole,
- 4,5-diamino-3-methyl-1-(3'-methoxyphenyl)pyrazole,
- 10 - 4,5-diamino-3-methyl-1-(4'-methoxyphenyl)pyrazole,
- 3-aminomethyl-4,5-diamino-1-methylpyrazole,
- 3-aminomethyl-4,5-diamino-1-ethylpyrazole,
- 3-aminomethyl-4,5-diamino-1-isopropylpyrazole,
- 3-aminomethyl-4,5-diamino-1-tert-butylpyrazole,
- 15 - 4,5-diamino-3-dimethylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-dimethylaminomethyl-1-isopropylpyrazole,
- 4,5-diamino-3-dimethylaminomethyl-1-tert-butylpyrazole,
- 20 - 4,5-diamino-3-ethylaminomethyl-1-methylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-ethylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-isopropylpyrazole,
- 4,5-diamino-3-ethylaminomethyl-1-tert-butylpyrazole,
- 4,5-diamino-3-methylaminomethyl-1-methylpyrazole,
- 25 - 4,5-diamino-3-methylaminomethyl-1-isopropylpyrazole,
- 4,5-diamino-1-ethyl-3-methylaminomethylpyrazole,

- 1-tert-butyl-4,5-diamino-3-methylaminomethylpyrazole,
- 4,5-diamino-3-[( $\beta$ -hydroxyethyl)aminomethyl]-1-methylpyrazole,
- 4,5-diamino-3-[( $\beta$ -hydroxyethyl)aminomethyl]-1-isopropylpyrazole,
- 4,5-diamino-1-ethyl-3-[( $\beta$ -hydroxyethyl)aminomethyl]pyrazole,
- 1-tert-butyl-4,5-diamino-3-[( $\beta$ -hydroxyethyl)aminomethyl]pyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1,3-dimethylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-isopropyl-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-ethyl-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-tert-butyl-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-phenyl-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(2-methoxyphenyl)-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(3-methoxyphenyl)-3-methylpyrazole,
- 4-amino-5-( $\beta$ -hydroxyethyl)amino-1-(4-methoxyphenyl)-3-methylpyrazole,

- 4-amino-5-( $\beta$ -hydroxyethyl) amino-1-benzyl-3-methylpyrazole,  
- 4-amino-1-ethyl-3-methyl-5-methylaminopyrazole,  
- 4-amino-1-tert-butyl-3-methyl-5-methylaminopyrazole,  
5 - 4,5-diamino-1,3-dimethylpyrazole,  
- 4,5-diamino-3-tert-butyl-1-methylpyrazole,  
- 4,5-diamino-1-tert-butyl-3-methylpyrazole,  
- 4,5-diamino-1-methyl-3-phenylpyrazole,  
- 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-methylpyrazole,  
10 - 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-phenylpyrazole,  
- 4,5-diamino-1-methyl-3-(2'-chlorophenyl)pyrazole,  
- 4,5-diamino-1-methyl-3-(4'-chlorophenyl)pyrazole,  
- 4,5-diamino-1-methyl-3-(3'-trifluoromethylphenyl)-pyrazole,  
15 - 4,5-diamino-1,3-diphenylpyrazole,  
- 4,5-diamino-3-methyl-1-phenylpyrazole,  
- 4-amino-1,3-dimethyl-5-phenylaminopyrazole,  
- 4-amino-1-ethyl-3-methyl-5-phenylaminopyrazole,  
- 4-amino-1,3-dimethyl-5-methylaminopyrazole,  
20 - 4-amino-3-methyl-1-isopropyl-5-methylaminopyrazole,  
- 4-amino-3-isobutoxymethyl-1-methyl-5-methylamino-pyrazole,  
- 4-amino-3-methoxyethoxymethyl-1-methyl-5-methylamino-pyrazole,  
25 - 4-amino-3-hydroxymethyl-1-methyl-5-methylamino-pyrazole,

- 4-amino-1,3-diphenyl-5-phenylaminopyrazole,
- 4-amino-3-methyl-5-methylamino-1-phenylpyrazole,
- 4-amino-1,3-dimethyl-5-hydrazinopyrazole,
- 5-amino-3-methyl-4-methylamino-1-phenylpyrazole,
- 5 - 5-amino-1-methyl-4-(N,N-methylphenyl)amino-3-(4'-chlorophenyl)pyrazole,
- 5-amino-3-ethyl-1-methyl-4-(N,N-methylphenyl)amino-pyrazole,
- 5-amino-1-methyl-4-(N,N-methylphenyl)amino-3-
- 10 phenylpyrazole,
- 5-amino-3-ethyl-4-(N,N-methylphenyl)aminopyrazole,
- 5-amino-4-(N,N-methylphenyl)amino-3-phenylpyrazole,
- 5-amino-4-(N,N-methylphenyl)amino-3-(4'-methyl-phenyl)pyrazole,
- 15 - 5-amino-3-(4'-chlorophenyl)-4-(N,N-methylphenyl)-aminopyrazole,
- 5-amino-3-(4'-methoxyphenyl)-4-(N,N-methylphenyl)-aminopyrazole,
- 4-amino-5-methylamino-3-phenylpyrazole,
- 20 - 4-amino-5-ethylamino-3-phenylpyrazole,
- 4-amino-5-ethylamino-3-(4'-methylphenyl)pyrazole,
- 4-amino-3-phenyl-5-propylaminopyrazole,
- 4-amino-5-butylamino-3-phenylpyrazole,
- 4-amino-3-phenyl-5-phenylaminopyrazole,
- 25 - 4-amino-5-benzylamino-3-phenylpyrazole,
- 4-amino-5-(4'-chlorophenyl)amino-3-phenylpyrazole,



- 4-amino-3-(4'-chlorophenyl)-5-phenylaminopyrazole,
  - 4-amino-3-(4'-methoxyphenyl)-5-phenylaminopyrazole,
  - 1-(4'-chlorobenzyl)-4,5-diamino-3-methylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole,
  - 5 - 4-amino-1-ethyl-3-methyl-5-methylaminopyrazole,
  - 4-amino-5-(2'-aminoethyl)amino-1,3-dimethylpyrazole,
- and the addition salts thereof with an acid.

7. Composition according to Claim 6,  
characterized in that the diaminopyrazoles of formula
- 10 (III) are chosen from:
- 4,5-diamino-1,3-dimethylpyrazole,
  - 4,5-diamino-3-methyl-1-phenylpyrazole,
  - 4,5-diamino-1-methyl-3-phenylpyrazole,
  - 4-amino-1,3-dimethyl-5-hydrazinopyrazole,
  - 15 - 1-benzyl-4,5-diamino-3-methylpyrazole,
  - 4,5-diamino-3-tert-butyl-1-methylpyrazole,
  - 4,5-diamino-1-tert-butyl-3-methylpyrazole,
  - 4,5-diamino-1-( $\beta$ -hydroxyethyl)-3-methylpyrazole,
  - 4,5-diamino-1-ethyl-3-methylpyrazole,
  - 20 - 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole,
  - 4,5-diamino-1-ethyl-3-hydroxymethylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-methylpyrazole,
  - 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole,
  - 4,5-diamino-3-methyl-1-isopropylpyrazole,
  - 25 - 4-amino-5-(2'-aminoethyl)amino-1,3-dimethylpyrazole,
- and the addition salts thereof with an acid.

8. Composition according to Claim 4 characterized in that the triaminopyrazoles of formula (IV) are chosen from 3,4,5-triaminopyrazole, 1-methyl-3,4,5-triaminopyrazole, 3,5-diamino-1-methyl-4-methylaminopyrazole and 3,5-diamino-4-( $\beta$ -hydroxyethyl)amino-1-methylpyrazole, and the addition salts thereof with an acid.

9. Composition according to any one of the preceding claims, characterized in that the diaminopyrazole(s) and/or the triaminopyrazole(s) and/or the corresponding addition salt(s) with an acid represent(s) from 0.0005 to 12% by weight relative to the total weight of the dye composition.

10. Composition according to Claim 9, characterized in that the diaminopyrazole(s) and/or the triaminopyrazole(s) and/or the corresponding addition salt(s) with an acid represent(s) from 0.005 to 6% by weight relative to the total weight of the dye composition.

11. Composition according to any one of the preceding claims, characterized in that the halogenated meta-aminophenol(s) of formula (I) and/or the corresponding addition salt(s) with an acid represent(s) from 0.0001 to 5% by weight relative to the total weight of the dye composition.

16. Composition according to any one of the preceding claims, characterized in that it is in the form of liquids, creams or gels or in any other form which is suitable for dyeing keratin fibres, and in particular human hair.

17. Process for dyeing keratin fibres, and in particular human keratin fibres such as the hair, characterized in that at least one dye composition as defined in any one of Claims 1 to 16 is applied to these fibres, and in that the colour is developed at acidic, neutral or alkaline pH with the aid of an oxidizing agent which is added to the dye composition just at the time of use, or which is present in an oxidizing composition that is applied simultaneously or sequentially.

18. Process according to Claim 17, characterized in that the oxidizing agent present in the oxidizing composition is chosen from hydrogen peroxide, urea peroxide, alkali metal bromates, persalts such as perborates, percarbonates and persulphates, and peracids.

19. Multi-compartment device or multi-compartment dyeing "kit", a first compartment of which contains a dye composition as defined in any one of Claims 1 to 16, and a second compartment of which contains an oxidizing composition.

# ABSTRACT

COMPOSITION FOR THE OXIDATION DYEING OF KERATIN FIBRES,  
COMPRISING A DIAMINOPYRAZOLE OR A TRIAMINOPYRAZOLE AND  
A HALOGENATED META-AMINOPHENOL, AND DYEING PROCESS

The present invention relates to a composition for the oxidation dyeing of keratin fibres, in particular human keratin fibres such as the hair, comprising at least one oxidation base chosen from diaminopyrazoles and triaminopyrazoles, in combination with at least one meta-aminophenol which is halogenated ortho to the phenol, as coupler, and to the dyeing process using this composition with an oxidizing agent.

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# Declaration and Power of Attorney for Patent Application

## Déclaration et Pouvoir pour Demand de Brevet

### French Language Declaration

En tant que l'inventeur nommé ci-après, je déclare par le présent acte que:

Mon domicile, mon adresse postale et ma nationalité sont ceux figurant ci-dessous à côté de mon nom.

Je crois être le premier inventeur original et unique (si un seul nom est mentionné ci-dessous), ou l'un des premiers co-inventeurs originaux (si plusieurs noms sont mentionnés ci-dessous) de l'objet revendiqué, pour lequel une demande de brevet a été déposée concernant l'invention intitulée

et dont la description est fournie ci-joint à moins que la case suivante n'ait été cochée:

☒ a été déposée le \_\_\_\_\_  
sous le numéro de demande des Etats-Unis ou le  
numéro de demande internationale PCT  
\_\_\_\_\_ et modifiée  
\_\_\_\_\_ (les cas échéant).

Je déclare par le présent acte avoir passé en revue et compris le contenu de la description ci-dessus, revendications comprises, telles que modifiées par toute modification dont il aura été fait référence ci-dessus.

Je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

DYEING COMPOSITION FOR KERATIN FIBRES

the specification of which is attached hereto unless the following box is checked:

☒ was filed on July 20, 1998 as United States Application Number or PCT International Application Number PCT/FR98/01591 and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

**French Language Declaration**

Je revendique par le présent acte avoir la priorité étrangère, en vertu du Titre 35, § 119(a)-(d) ou § 365(b) du Code des Etats-Unis, sur toute demande étrangère de brevet ou certificat d'inventeur ou, en vertu du Titre 35, § 365(a) du même Code, sur toute demande internationale PCT désignant au moins un pays autre que les Etats-Unis et figurant ci-dessous et, en cochant la case, j'ai aussi indiqué ci-dessous toute demande étrangère de brevet, tout certificat d'inventeur ou toute demande internationale PCT ayant une date de dépôt précédant celle de la demande à propos de laquelle une priorité est revendiquée.

Prior foreign application(s)  
Demande(s) de brevet antérieure(s)

Priority Not Claimed  
Droit de priorité non revendiqué

|          |           |
|----------|-----------|
| 97/10857 | France    |
| (Number) | (Country) |
| (Numéro) | (Pays)    |
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| (Number) | (Country) |
| (Numéro) | (Pays)    |

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| 1 September 1997          | <input type="checkbox"/> |
| (Day/Month/Year Filed)    |                          |
| (Jour/Mois/Anné de dépôt) |                          |
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| (Day/Month/Year Filed)    | <input type="checkbox"/> |
| (Jour/Mois/Anné de dépôt) |                          |

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 119(e) du Code des Etats-Unis, de toute demande de brevet provisoire effectuée aux Etats-Unis et figurant ci-dessous.

|                   |                 |
|-------------------|-----------------|
| (Application No.) | (Filing Date)   |
| (N° de demande)   | (Date de dépôt) |
| <hr/>             |                 |
| (Application No.) | (Filing Date)   |
| (N° de demande)   | (Date de dépôt) |

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 120 du Code des Etats-Unis, de toute demande de brevet effectuée aux Etats-Unis, ou en vertu du Titre 35, § 365(c) du même Code, de toute demande internationale PCT désignant les Etats-Unis et figurant ci-dessous et, dans la mesure où l'objet de chacune des revendications de cette demande de brevet n'est pas divulgué dans la demande antérieure américaine ou internationale PCT, en vertu des dispositions du premier paragraphe du Titre 35, § 112 du Code des Etats-Unis, je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations, dont laquelle est devenue disponible entre la date de dépôt de la demande antérieure, et la date de dépôt de la demande nationale ou internationale PCT de la présente demande:

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International Application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose any or all information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

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| (N° de demande)   | (Date de dépôt) |
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| (Application No.) | (Filing Date)   |
| (N° de demande)   | (Date de dépôt) |

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| (Status) (patented, pending, abandoned)          |  |
| (Status) (breveté, en cours d'examen, abandonné) |  |
| <hr/>  |  |
| (Status) (patented, pending, abandoned)          |  |
| (Status) (breveté, en cours d'examen, abandonné) |  |

Je déclare par le présent acte que toute déclaration ci-incluse est, à ma connaissance, véridique et que toute déclaration formulée à partir de renseignements ou de suppositions est tenue pour véridique; et de plus, que toutes ces déclarations ont été formulées en sachant que toute fausse déclaration volontaire ou son équivalent est passible d'une amende ou d'une incarcération, ou des deux, en vertu de la Section 1001 du Titre 18 du Code des Etats-Unis, et que de telles déclarations volontairement fausses risquent de compromettre la validité de la demande de brevet ou du brevet délivré à partir de celle-ci.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

**French Language Declaration**

POUVOIRS: En tant que l'inventeur cité, je désigne par la présente l'(les) avocat(s) et/ou agent(s) suivant(s) pour qu'ils poursuive(nt) la procédure de cette demande de brevet et traite(nt) toute affaire s'y rapportant avec L'Office des brevets et des marques: (*mentionner le nom et le numéro d'enregistrement*).

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this patent application and transact all business in the Patent and Trademark Office connected therewith: (*list name and registration number*):

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| Signature du second inventeur                       | Date   | Second Inventor's signature<br><u>Marie-Pascale AUDOUSSET</u>                 | Date<br><u>03/20/2000</u>                 |
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| Signature d'inventeur                               | Date   | Third Inventor's signature  | Date                                      |
| Domicile  | Residence  |   |   |
| Nationalité:  | Citizenship  |   |   |
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